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Effects of Propane/Natural Gas Blended Fuels on Gas Turbine Pollutant Emissions

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U.S. natural gas composition is expected to be more variable in the future. Liquefied natural gas (LNG) imports to the U.S. are expected to grow significantly over the next 10-15 years. Unconventional gas supplies, like coal-bed methane, are also expected to grow. As a result of these anticipated changes, the composition of fuel sources may vary significantly from existing domestic natural gas supplies. To allow the greatest use of gas supplies, end-use equipment should be able to accommodate the widest possible gas composition. For this reason, the effect of gas composition on combustion behavior is of interest. This paper will examine the effects of fuel variability on pollutant emissions for premixed gas turbine conditions. The experimental data presented in this paper have been collected from a pressurized single injector combustion test rig at the National Energy Technology Laboratory (NETL). The tests are conducted at 7.5 atm with a 589K air preheat. A propane blending facility is used to vary the Wobbe Index of the site natural gas. The results indicate that propane addition of about five (vol.) percent does not lead to a significant change in the observed NOx emissions. These results vary from data reported in the literature for some engine applications and potential reasons for these differences are discussed.

1. Introduction

Over the next 15-20 years, projections indicate that domestic natural gas production will be outpaced by demand. As a result, Liquefied Natural Gas (LNG) imports to the United States are projected to increase over this same period. In 2005, LNG imports amounted to about 3% of the total domestic gas supply. However, by 2025 projections from the Energy Information Administration indicate that LNG imports could account for roughly 15% of the domestic U.S. gas supply[1].

Since shipping costs for LNG are an important consideration, attempts to manage these costs can lead to differences in the chemical composition of LNG relative to domestic natural gas. For example, LNG typically has lower concentrations of inert gases, such as nitrogen and carbon dioxide, since these constituents do not add energy value to the LNG product. In fact, these inert constituents will be removed by processing and boil-off during transportation.[2] In addition, LNG fuel compositions typically have a higher concentration of heavier hydrocarbons.

Fuel variability is of particular concern for land-based gas turbines that utilize lean premixed combustion technology. The sensitivity of some gas turbine combustors to changing operating conditions is not widely appreciated. In some engines, ambient temperature changes can lead to damaging oscillations[3]. However, engines can be adjusted, or tailored, to operate on widely different fuel composition. Kurz[4] notes that some engines may allow as much as a 10% variation in fuel heating value, while others can accommodate less than 2-3%. Although engines may be adjusted for different fuels, a key question is whether engines that have been designed to meet emissions standards on domestic natural gas can accommodate sudden changes in fuel composition, without causing machine upsets, excess emissions, or component damage.

For premixed gas turbine applications, only a limited amount of public information is available that describes the effects of gaseous fuel variability on pollutant (i.e., NOx and CO) emissions. Lee[5] describes the effect of methane, ethane, propane, and other hydrocarbon fuels on NOx emissions in a premixed jet-stirred reactor flame. These data are collected at atmospheric pressure, but at firing temperatures that represent modern gas turbines (1790K = 2762F). Heat loss effects are managed by controlling the exit temperature of the JSR. For a 2.3 millisecond residence time, the NOx emissions increase modestly (i.e., a difference of about one part-per-million on a 5 ppm baseline) as the fuel is varied from pure methane to pure propane. Noting that variations in the higher hydrocarbon concentrations expected for existing gas specifications will be much smaller than differences studied by Lee[5], one would expect smaller changes in NOx emissions. However, kinetic modeling predicts otherwise.

Klassen[6] uses chemical reactor modeling (CRM) to predict the effect of natural gas composition on NOx emissions. Although no experimental validation of this data is presented, the model shows that NOx levels can vary by as much as a factor-of-two for a constant Wobbe Index and constant flame temperature. It is emphasized that these are calculations from available kinetic models that have been tuned to engine-specific conditions. As discussed by Klassen[6], they should be subject to experimental validation.

Flores et al.[7] have experimentally investigated the emissions from a premix, swirl-stabilized combustor, using fuel blends of natural gas with as much as 15% ethane and 20% propane. The results are limited to atmospheric pressure, but realistic air-preheat temperatures are used. This work concluded that fuel composition has a significant effect on NOx emissions, but a subtle dependency on the fuel premixing approach was also observed. These tests were performed at a constant equivalence ratio, so the flame temperature may have varied slightly with fuel composition. In an earlier paper, Flores et al.[8] show the observed NOx emissions as a function of flame temperature from a nearly identical test rig. For flame temperatures below 1850K, fuel blends of 80 percent methane/20 percent propane produce negligible changes in the observed NOx emissions relative to 100 percent methane. However, for flame temperatures above 1850K, the fuel composition shows an effect.

Hack and McDonell[9] have tested a recuperated 60kW micro-turbine, using fuel blends with 77 – 100% methane, 0-16% ethane and 0 – 20% propane. These studies have been conducted at fixed engine load and constant turbine exit temperature. These tests show a significant fuel composition effect on NOx emissions. It is interesting to note that two different test series using two different micro-turbine generators are discussed in Hack and McDonell[9]. One of these data sets was also described by McDonell and Kay[10] in 2003. In this earlier work, the qualitative effects of fuel composition on pollutant emissions are clearly shown in the time-series data, but the day-to-day variations in NOx measurements are as large as the variations observed as a result of changing fuel composition.

Limited data from commercial gas turbine installations are available. Nord and Anderson[11,12] have studied operating engine data using lean premixed and diffusion-style combustion technologies. They have shown that normal day-to-day variations in fuel composition had limited effect on the emissions, but larger, sudden changes in the fuel composition could have a significant impact on combustor performance.

In another studied funded by the California Energy Commission,[13] four turbine installation along a Pacific Gas and Electric natural gas pipeline were monitored during an excursion in fuel quality. Although there is a lot of scatter in the process data, the general trend that NOx emissions upstream of the exhaust after-treatment devices were slightly higher during this event. In all instances, the emissions downstream of the exhaust after-treatment devices did not change as a result of changes in the fuel composition.

In response to growing interest on the relation between gas composition and power generation, a recent study has been completed at the U.S. Department of Energy's National Energy Technology Laboratory. The purpose of this study is to investigate gas interchangeability effects which covered a broad range of issues, including combustion instabilities.[14,15] The purpose of this paper is to address the effect of fuel composition on NOx emissions under well-controlled gas turbine operating conditions. In the following sections, the approach and experimental apparatus is described. Preliminary results from these studies are then presented, followed by a summary of the results and conclusions.

2. Approach

In lieu of conducting large-scale emission testing on specific engine configurations, a more generic approach has been pursued. The effect of fuel composition on pollutant emissions will be investigated using an existing single-injector pressurized combustion rig at the NETL. This test rig is capable of studying combustion instabilities, as well as, characterizing the pollutant emissions as a function of fuel composition. The test rig is designed to operate at pressures as high as 10 atmospheres with an air flow rate of 0.75 kg/s (1.6 lb/s). The air preheat temperature is limited to 589K (600F). The fuel composition is varied by blending propane fuel with pipeline natural gas at concentrations of about 5 volume percent. This level of propane blending has been chosen to achieve Wobbe Index values that are higher than typical pipeline natural gas, but still within the projected range of most LNG fuel sources. A two-factor, two-level factorial design with replications at a centerpoint will be used to assess the effects of fuel composition on pollutant emissions. The experimental apparatus will be described in more detail in the following paragraphs.

3. Fuel Blending System

In order to simulate the fuel composition of an imported LNG fuel, NETL's existing high-pressure gas turbine combustion facility has been modified. Several options for simulating LNG fuel have been evaluated, and ultimately a propane blending facility has been constructed (see Fig. 1). The pressure of the propane vapor is controlled by a closed-loop water heating unit that externally heats a pressure vessel partially filled with commercial grade propane. In accordance with the National Fire Protection Association Code that addresses Liquefied Petroleum Gases (NFPA-58), the gases are mixed in an area that meets Class I, Division 2 electrical classification. The blended gas, having a density that is lighter-than-air, does not have to meet the stringent requirements of NFPA-58.

The propane blending unit is shown in Figure 1. The insulated tank on the left is a double-walled pressure vessel. Water circulates through the outer volume to heat the inner volume containing propane. As soon as the propane vapor exits the top of the tank, electrical heat tracing is used to prevent condensation in the process lines. The propane vapor is split into two metered streams. The larger of these two streams is blended with the main premix fuel. Both the main propane and the main natural gas flows are metered independently using Emerson-Rosemount 3095 multivariable mass flow transmitters. The second propane stream is mixed with the natural gas pilot fuel. Both of these pilot fuel streams are metered independently using Siemens (SITRANS F C Massflo Mass 2100 DI 1.5) coriolis meters.

Figure 1: Photo of Propane Blending Station

The dew-points of the blended gas streams are low enough that heat-tracing is no longer required after the gases are mixed. The gas composition in the blended gas streams are monitored using an on-line gas chromatograph (Agilent 3000 Micro-GC). This instrument has also been used to analyze the gas composition of the propane prior to start-up.

It is important to note that the as-received propane is not pure (see Fig. 2a). Initially, ethane concentrations in the vapor phase ranged from 10-15 percent (volume basis). However, due to the significant differences in the volatility of propane and ethane, the ethane vaporizes more readily and is preferentially consumed. As a result, after about two-hours of testing, the concentration of ethane in the vapor phase decreases to less than 2 percent of the fuel (see Fig 2b). These changes in fuel composition complicate the flow metering, the stoichiometry of the combustion, and potentially the interpretation of the data.

4. Combustion Test Rig

The combustion rig, instrumentation, and data acquisition are similar to the setup described in Chorpening et al.[16] A cross-section of the test rig is shown in Fig. 3. Preheated air enters the plenum region from metered high temperature flow loops. All flow measurements (fuel and air) are within two-percent of flow standards. Fuel and air are mixed inside the premix nozzle prior to entering the 19.3 cm (7.6 in) diameter combustion chamber. The walls of the combustion chamber are water-cooled and provide strong acoustic feedback. The removable refractory insert shown in Fig. 3 can be re-located to alter the acoustic characteristics of the test rig. In these tests, this refractory plug has been located to make the length of the combustion zone 0.91 m (36 in).

A near-commercial premix fuel injector will be used in these studies as described by Benson et al.,[17] and Chorpening et al.[16]. Pilot fuel is injected through the centerbody tip via 12 holes (see Fig. 4). In addition to providing a means for flame stabilization, this fuel flow also provides cooling to the nozzle tip. The pilot fuel flow rate is maintained at five percent of the total fuel flow. When propane is blended with the main fuel flow, propane is also blended with the pilot fuel flow at the same concentration.

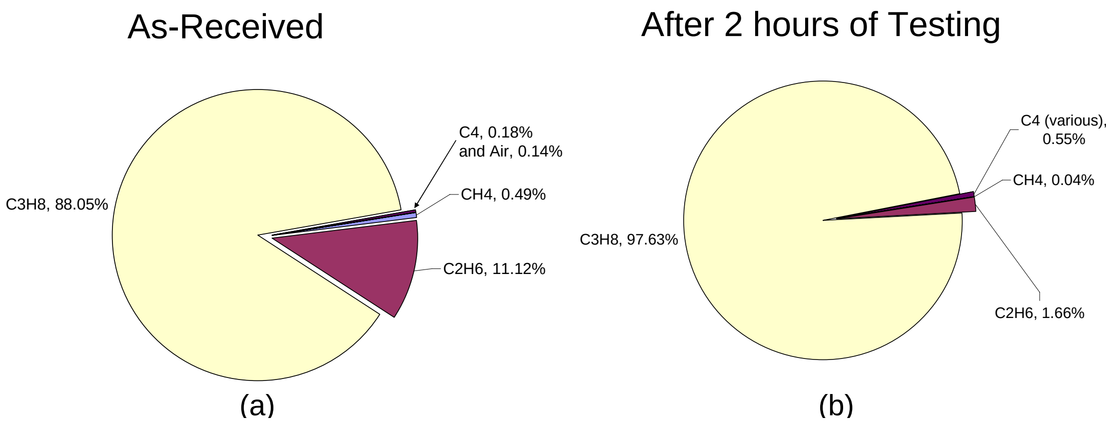

Figure 2: Propane Fuel Composition Variations

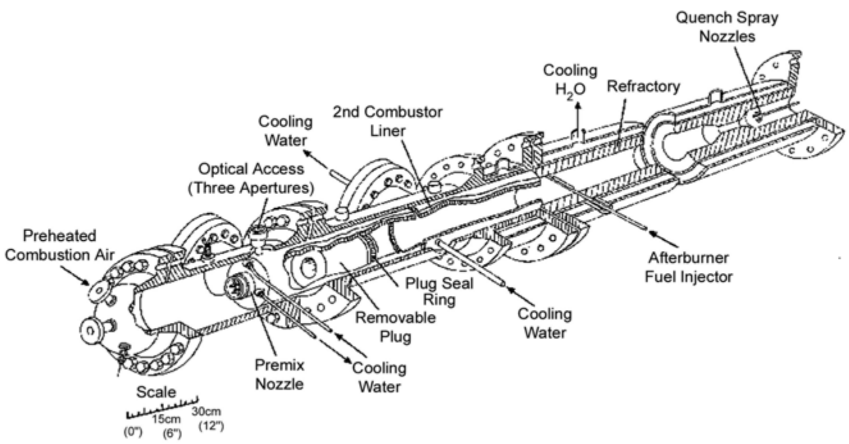

Figure 3: Cross-Section Of Experimental Combustion Test

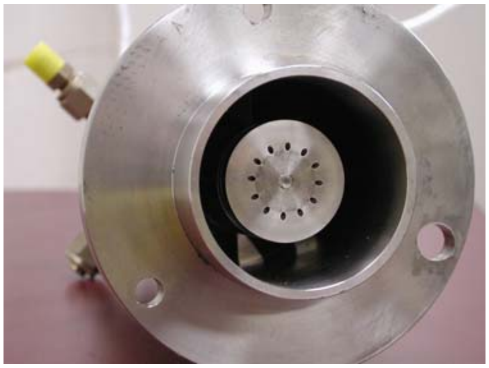

Figure 4: Premixer Photo

5. Gas Sampling System

A water-cooled stainless-steel sample probe is inserted into the combustor section approximately 0.53 meters (21-in) from the combustor inlet. This probe is located upstream of the refractory insert described earlier, and extracts an area-weighted gas sample through three 1.85 mm (0.073-in) diameter holes. Since the test rig operates at elevated pressure, the gas sample flows through the holes in the probe to the sampling system.

Condensate is particularly important to avoid for single-digit NOx measurements, due to the solubility of NO_2 in water which could bias the results. The gas sample is transported through a heated 6.4 mm (1/4-in) stainless-steel line to a pressure control and sample conditioning station (see Fig. 5). A small pressure control valve is used to vent excess flow though the sampling system and maintain a constant pressure in the gas analyzer manifold. Some of the gas analyzers are sensitive to changes in flow or pressure, so this pressure control is finely tuned and closely monitored during testing. There are also a series of block valves that allowed the chiller and gas analyzers to be isolated, while high pressure nitrogen is used to back-flush the sampling system.

The exhaust sample is split with a portion of the heated sample passing through a NO_2-NO converter prior to entering a chiller/dryer. Conversion of water soluble NO_2 to non-soluble NO ensures a more accurate NOx measurement. This leg of the flow loop also supplies sample to the O_2 analyzer. A separate flow loop through the chiller/dryer that does not pass through the converter supplies the CO and CO_2 measurement.

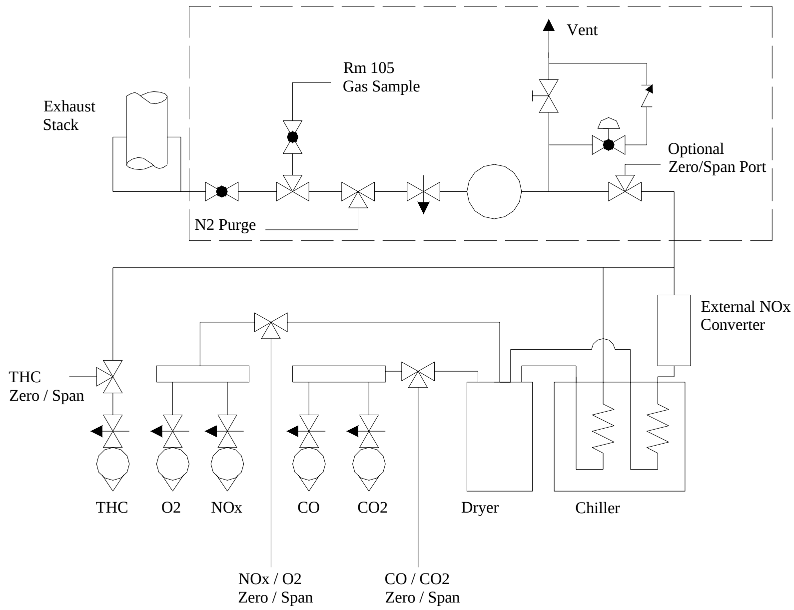

Figure 5: Schematic of Gas Sampling Process

6. Results

Before the results are discussed, it is important to note two things. First, the CO concentrations in the exhaust are less than one part-per-million for all of the conditions investigated. Therefore, the CO emission results will not be presented.

Secondly, combustion-induced pressure oscillations can significantly confound the measurements of pollutant emissions (see Fig 6). The distinct change in the RMS pressure level shown in Fig. 6 leads to a change in NOx emissions of almost 40-50%. It is believed that mixing is enhanced when the combustion is unstable and the improved mixing leads to lower NOx production. This type of behavior is often observed in rig tests and must be considered when investigating the effects of fuel composition on pollutant emissions. The data shown in Fig. 6 has been collected during initial exploratory studies using pipeline natural gas. These exploratory studies are conducted to determine the location of the dynamic stability boundaries. Subsequent emissions tests can then be performed in a region in which pressure oscillations do not confound the experiments (i.e., for equivalence ratio conditions less than 0.55). For the range of fuel compositions investigated in this study, this stability boundary did not change significantly. The effect of fuel composition on combustion instabilities has also been investigated at NETL and this topic is described in a separate paper[15].

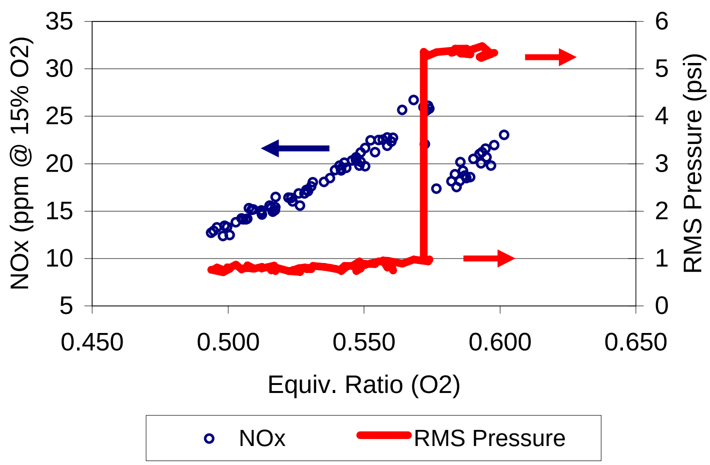

Figure 6: Potential Confounding Effect of Combustion Instabilities and NOx Emissions

The effects of propane concentration in the fuel and the fuel-air equivalence ratio are investigated using a statistically designed experiment. The combustor pressure is fixed at 760 kPa (110 psia), and the inlet-air temperature is fixed at 589K (600F). The equivalence ratio range is selected to achieve RMS pressure levels that are less than one percent of the mean operating pressure, or less than 7.5 kPa (1.1 psi). As shown in Table 1, the fuel-air equivalence ratio is varied from 0.44 to 0.52. The propane level is varied from approximately zero (i.e., pipeline natural gas) to a nominal value of about five percent.

It should be noted that the pipeline natural gas composition changed slightly over the course of this experiment (see Fig. 7), and the on-line gas chromatograph analyses are used to quantify these changes. The propane concentration in the baseline natural gas shifted from about 0.7 percent to about 1.0 percent, and the Wobbe Index increased from about 1360 to 1380 BTU/scf. It is believed that this shift in baseline fuel occurred somewhere between Test Point 3 and Test Point 4. This change in the baseline fuel may have increased the observed scatter in the experimental data, but it is believed that this did not affect our conclusions, due to the randomization of the test points. However, additional tests are planned to further verify the data presented here.

Figure 8 shows the measured NOx emissions plotted as a function of the propane concentration in the fuel. The magnitudes of the observed NOx emissions are representative of a partially premixed (i.e., diffusion piloted) gas turbine combustor. The replications at a propane level of nearly four percent show that the repeatability in the NOx measurements is on the order of 0.9 parts-per-million, assuming a 95% confidence interval. Therefore, the differences observed between the baseline fuel and the propane blended-fuel is not statistically significant.

Table 1: Test Matrix

Test Point†	Target Equiv. Ratio	Actual Equiv. Ratio (flows)	Target Propane Level	Actual Propane Level (%)	Adiabatic Flame Temperature (K)
6	0.52	0.510 $\pm$ 0.005	5.0%	5.46 $\pm$ 0.002	1717
1	0.52	0.503 $\pm$ 0.004	0.0%	0.69 $\pm$ 0.095	1708
3	0.44	0.445 $\pm$ 0.004	5.0%	5.33 $\pm$ 0.029	1599
5	0.44	0.453 $\pm$ 0.005	0.0%	1.14 $\pm$ 0.016	1607
2	0.48	0.474 $\pm$ 0.005	3.5%	4.00 $\pm$ 0.018	1654
4	0.48	0.479 $\pm$ 0.005	3.5%	4.19 $\pm$ 0.015	1660
7	0.48	0.478 $\pm$ 0.005	3.5%	4.08 $\pm$ 0.001	1659
8	0.44	0.453 $\pm$ 0.005	0.0%	1.00 $\pm$ 0.001	1608

† The test points have been numbered in chronological order

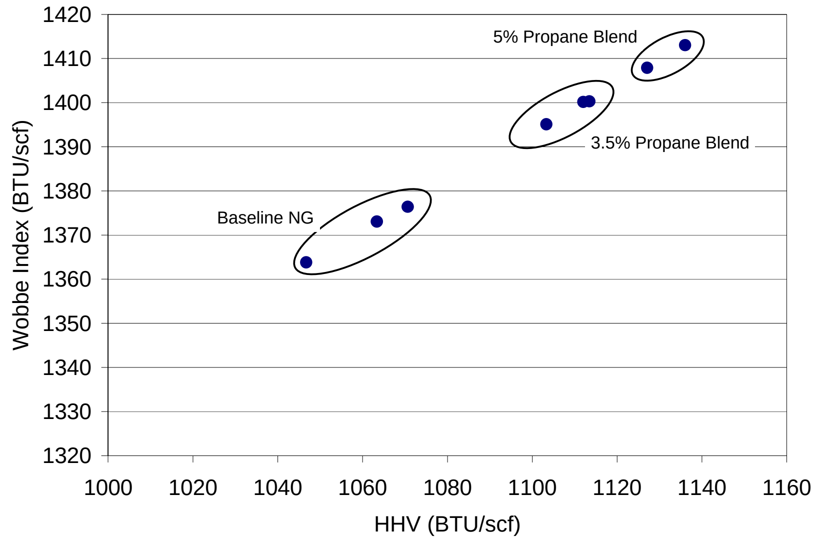

Figure 7: Wobbe Plot of Gas Compositions Investigated

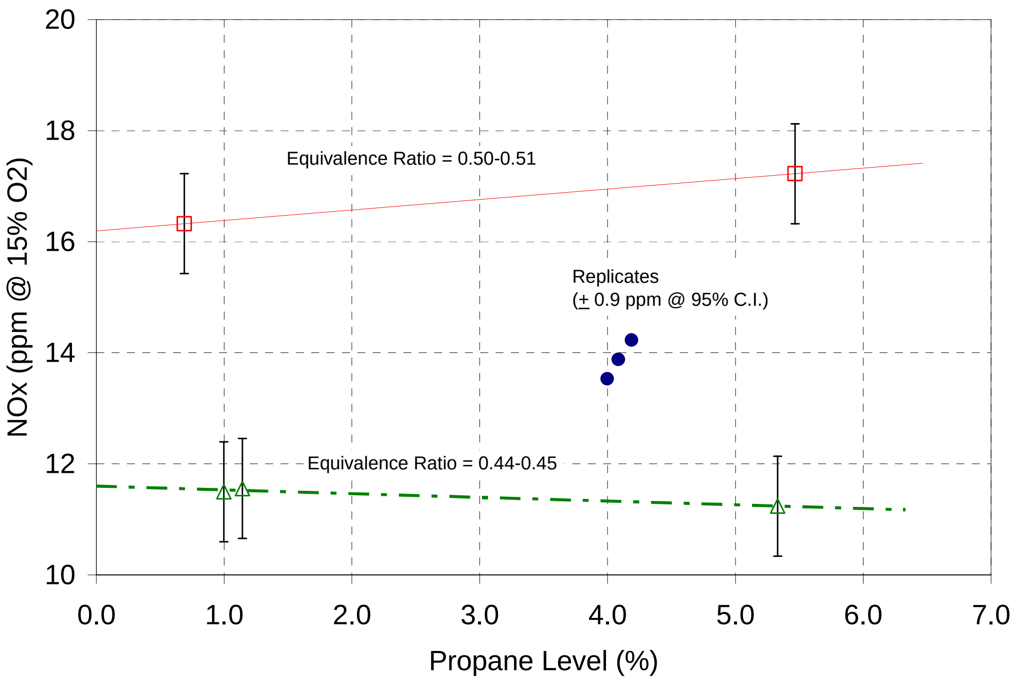

Figure 8: NOx Emissions as a Function of Propane Level

In an attempt to better understand the observed scatter in the data, it is recognized that the adiabatic flame temperature is a function of both the fuel-air equivalence ratio and the fuel composition. Therefore, adiabatic flame temperature calculations have been performed using Cantera.[18] The species thermodynamic information required for these calculations is derived from the GRI-Mech 3.0 kinetic mechanism[19] (see Table 1). Since GRI-Mech 3.0 does not include hydrocarbons heavier than propane, the hydrocarbon species heavier than propane have been neglected. The inert species in the fuel are included, but the concentrations of argon and carbon dioxide in the air have been neglected. It should be noted that localized temperatures in the flame region may be significantly higher than the values calculated, since the diffusion pilot fuel will burn at near-stoichiometric temperatures. However, for the purposes of this analysis, the pilot fuel has been combined with the premixed fuel and ideal mixing has been assumed.

By transforming the independent variables to flame temperature, a very good correlation is achieved (see Fig. 9). If the data grouped near a flame temperature of 1600K is considered more carefully, Table 1 shows that the actual equivalence ratio is slightly lower when propane is added, and hence the calculated flame temperature is lower. Although this difference is small, it reduces the flame temperature and the observed NOx emissions as shown in Fig. 9. For the data grouped between 1700-1725K, Table 1 shows that the actual fuel-air equivalence ratio is slightly higher when the propane is added. Again, the differences are small, but the flame temperature parameter seems to produce a good correlation of NOx emissions over the range of operating conditions investigated.

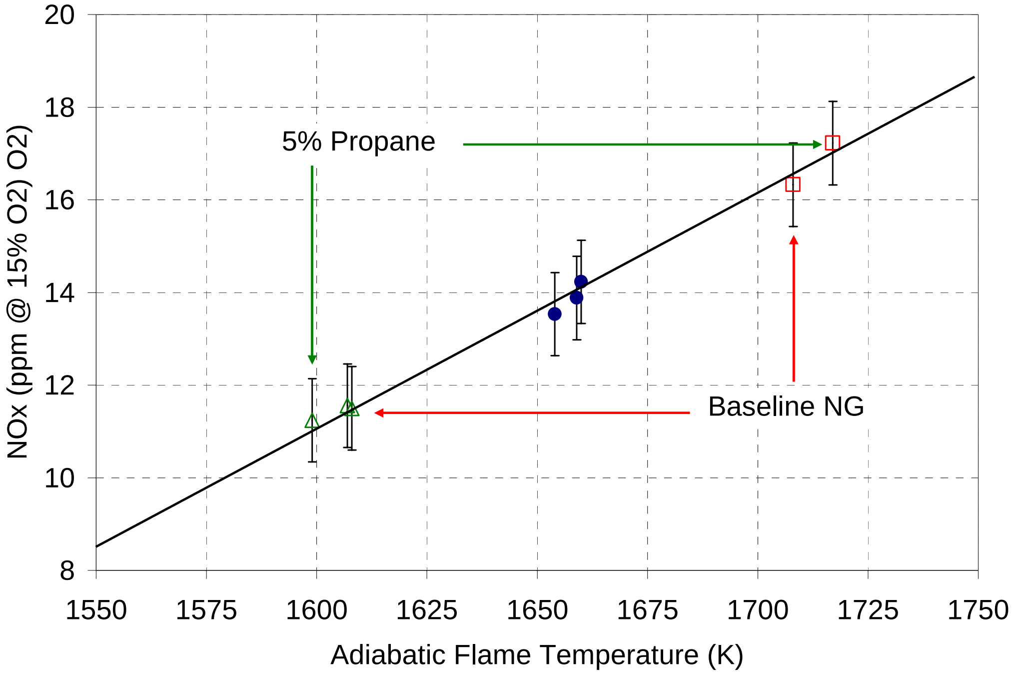

Figure 9: NOx Emissions as a Function of Flame Temperature

7. Discussion of Results

The results from these rig tests do not indicate a statistically significant change in NOx emissions as propane is blended with the pipeline natural gas at levels as high as five percent. However, other studies have observed a significant effect with propane addition. One important difference is the fact that the operating conditions in this test have been chosen to avoid confounding effects due to combustion instabilities. As a result, the operating conditions described in this paper cover a fairly low range of flame temperatures. It is also important to note that the fuel in this nozzle configuration is not 100% premixed. A pilot fuel flow (5% of total fuel) is used at all operating conditions, and this is the reason for the higher NOx levels observed in this test rig.

In an actual gas turbine engine operating at a constant power condition (i.e., a constant product of the mass flow rate and enthalpy), variations in the fuel composition may have a small effect on the mass flow rate of products and the specific heat of the turbine working fluid. A simple analysis has been conducted for four different fuel compositions, three of which have been taken from Hack and McDonell[9] (see Table 2). For all four cases, a constant flame temperature is initially assumed. Or in other words, the equivalence ratio is chosen to achieve the same adiabatic flame temperature for all three fuel compositions. Due to these slight changes in gas properties, it is estimated that the flame temperature may have to change 5-15°K in order to achieve the same power. Admittedly, this is a simplified analysis, but it is believed that for the range of fuel variability expected for LNG imported fuel, these effects will be small, and potentially insignificant.

One other point is worth noting from Table 2. The effect of diluent addition can have a significant effect on the amount of fuel required to achieve constant power output from the turbine. In fact, diluent addition of approximately 4-5 percent can actually lead to a lower flame temperature (i.e., 5-10%) in the combustor in order to achieve the same power output. This is consistent with the observation of Hack and McDonell[9] which showed that the NOx emissions actually decrease with the addition of inert species in the fuel.

Table 2: Summary Table of Fuel Composition Effects on Exhaust Gas Properties

		Case 1	Case 2	Case 3	Case 4
Fuel Components (mole-fraction)	CH_4	0.963	0.82	0.77	0.927
	C_2H_6	0.015	0.1575	0.0111	0.015
	C_3H_8	0.0033	0.0032	0.203	0.0033
	CO_2	0.141	0.0144	0.0109	0.015
	N_2	0.0	0.0	0.0	0.04
Adiabatic Flame Temperature (K)		1830	1830	1830	1830
Equivalence Ratio		0.520	0.517	0.513	0.547
Combustion Products Mass Flow Relative to Mass Flow of Combustion Air		1.027	1.027	1.027	1.031
Products of Combustion Specific Heat (J/kg K)		1369	1365	1360	1369

8. Summary and Conclusions

This effort has been pursued in response to growing interest in natural gas fuel variability. Available information regarding the effects of fuel composition on pollutant emissions is limited. The tests described in this paper have been collected from a pressurized combustion rig operating at 7.5 atmospheres with a 589K air preheat.

Pressurized testing with heavy hydrocarbons is not straightforward. Code requirements for handling pressurized propane are significant and should be considered carefully. Fuel purity and distillation effects can occur during testing, and on-line fuel composition analyses are essential for maintaining acceptable process control and data quality.

Combustion instabilities can also significantly affect the observed NOx emissions. The test rig used in this testing provides a quiet operating range in which the effect of fuel composition on emissions can be investigated without confounding the effects of combustion instabilities with fuel composition.

The following conclusions are supported by the data presented in this paper.

- The effect of propane blending to levels of about five volume percent in the fuel did not have a statistically significant effect on the observed NOx emissions.
- The fuel-air equivalence ratio did have a significant effect on the NOx emissions. The overall equivalence ratio was varied from 0.44 to 0.52, and five percent of the total fuel was introduced as a diffusion pilot.
- The NOx emission data presented in this paper show a strong correlation with adiabatic flame temperature.

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